THE USE OF GAMMA RADIATION TO MEASURE MOISTURE DISTRIBUTION DURING DRYING PROCESSES

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SUMMARY

A gamma ray attenuation method was developed to measure the transient moisture distribution in a hygroscopic material during a dehydration process. The gamma ray technique was applied to the freeze dehydration of meat to study the receding of the ice-vapor phase front and the moisture distribution in the dry and frozen sections of the meat. The measurement of the temperature distribution in the meat was made in conjunction with the moisture distribution in order to investigate the relation of the minimum temperature in the meat with phase front position.

The gamma ray method developed consisted of passing a gamma ray beam through the material in which the moisture content was to be determined and using a scintillation counting system to determine the radiation absorbed by the material. The change in absorption rate of the gamma ray was related to the change in moisture content of the material.

After the passage of the phase front, the test results indicate that there was no more than a three percent variation in the moisture content of the dried meat throughout the drying process. Also, there was less than a three percent change in the moisture content of the wet frozen section of the meat until the passage of the phase front.

The results of this work indicate that the temperature minimum occurs at the same point as the ice-vapor phase of the drying process. The process was conducted at three total chamber pressures and the results

of the tests indicate that a faster drying rate was obtained at one torr as compared to slower rates at two and three torr chamber pressure.

This test conducted on the freeze-drying of meat indicates that the gamma radiation technique may be applied to other types of drying processes to determine moisture distribution during drying. The moisture content was measured at a very small area in the sample, thus allowing the determination of moisture content to be considered as a discrete position in the sample.

NOMENCLATURE

English Letters		Units
A	atomic weight	Kg/mole
I	residual intensity of radiation beam	photon/cm^2-sec
I_{dr}	residual intensity of radiation beam through dry product	photon/cm^2-sec
I_o	intensity before attenuation	photon/cm^2-sec
I_w	residual intensity of radiation beam through wet product	photon/cm^2-sec
K	linear attenuation coefficient for pair-production	cm^{-1}
k	experimental constant for calibration of radiation beam to moisture content	cm^3/gram
N_a	Avogadro's number	atoms/Kg-mole
P	total chamber pressure	torr(mm of Hg)
T	time	hour
u	moisture content	gram/cm^3
x	thickness	cm
Z	atomic number	protons/atom
Greek Letters		
γ	linear attenuation coefficient for photoelectric effect	cm^{-1}
$_e\mu$	mass attenuation coefficient	cm^2/electron
μ_o	total linear attenuation coefficient	cm^{-1}
ρ	density of absorber	g/cm^3

Greek Letters		Units
ρ_{dr}	density of dry absorber	g/cm^3
σ	linear attenuation coefficient for Compton process	cm^{-1}

CHAPTER I

INTRODUCTION

Statement of Intent

It is the intention of this study to design and develop a method for measuring local moisture content during drying processes. The method developed will be demonstrated on a freeze-drying process using meat.

Local Moisture Measurement During Drying Processes

Current industrial drying processes are very extensive; due to economic considerations, it is very desirable to improve present drying techniques and equipment. More thorough experimental investigations of drying processes would be extremely desirable in order to aid in the development of mathematical models which could be applied to drying processes. Most studies concerned with drying consider only total drying times instead of a more detailed investigation of the transient drying conditions. It is desirable to examine the local moisture content throughout a body during drying processes.

This determination of local moisture must be conducted in such a manner that it will not affect the mass flow out of the product, while at the same time giving an accurate measurement of the moisture content in the product. It is important to measure the moisture in a small portion of the body so that the measurement will apply to a discrete position in the body.

There are two types of moisture contained in a hygroscopic material,

chemical bound and free moisture that is entrained between the fibers. Since both types of moisture are involved in drying processes, it is a requirement of local moisture measurement that both be measured. Thus we have the three requirements of a local moisture measurement in a hygroscopic material:

1. The measurement will not affect the normal drying process.
2. The area of measurement should be small.
3. Both types of moisture in the material must be measured.

Moisture Measurement Devices

There have been several methods used to measure local moisture content during the drying of a hygroscopic material. The following discussion will give a general survey of moisture measuring techniques.

Resistance Method

This method consists of placing two probes into a sample and measuring the electrical resistance between them. Hardacker and Rawcliffe (1)* have shown that this method works well at low moisture content (0-14 percent based on dry sample weight); however, as the moisture content increases there is less dependence of resistance on moisture content until between 30 and 100 percent the resistance is practically independent of moisture content. Also, the range of resistance determinations is so large (1 ohm to 1000 meg ohms), that it would be difficult to find an accurate and simple instrument to measure it. Measurements made with this method involve rather sizable portions of the product. Therefore, it is difficult to use this method for measuring moisture content in small regions.

*Numbers in parentheses refer to references in the literature cited section.

Capacitance Method

The capacitance method consists of measuring the change in dielectric constant with moisture content of a material placed between two plates. Vitins (2) has shown that this method requires that there be no change in the amount of the product between the plates. The change in the dielectric must relate only to changes in moisture content to give proper readings. This method also requires that as the moisture content increases, the frequency of the current used to measure the dielectric must increase and, if the frequency is held constant, then at high moisture content the capacitance does not depend on moisture content. Also, there is always danger of electrical discharge of the capacitor if the moisture content becomes very high and the probes are not properly insulated.

Electrolytic Method

Basically this device consists of a probe that is a non-conductor which is coated with a conducting hydroscopic salt. The probe is then inserted into the product and the electrical resistance of the salt is a function of the moisture absorbed by the salt. Since Vitins (2) and McLeod (3) have shown the resistance of the salt is a function of the pressure and temperature as well as moisture, this requires the measurement of these to effectively calibrate the instrument. This method only measures the entrained moisture in the fibers and not the bound water in the cells. The requirement for equilibrium to be reached between the salt and the moisture injects a lag time which prohibits short readings (a minute or less).

X-Ray Technique

The x-ray method consists of passing an x-ray beam through the sample and measuring the attenuation of the beam. The attenuation can be related to the density of the sample and thus to the changing moisture content. Since x-rays are a very low energy photon, the absorption rate is a function of the thickness of the material as shown by Evans (4); thus, a slight change in the thickness of the material seen by the beam would have a large effect on the absorption. Therefore, it is difficult to relate the attenuation to moisture content. Also, the equipment is very expensive compared to similar techniques using higher energy photons and charged particles

Charged Particles and Gamma-Ray Beams

This method is similar to the x-ray technique in that a beam is passed through the body in which moisture is to be measured. The attenuation of the beam is measured and related to moisture content of the body. However, the energies of these rays are much higher than x-rays. The different beams that may be used consist of neutrons or alphas or betas or gamma rays.

The neutron has an affinity to the hydrogen atom, therefore, it has been shown by Belcher et al. (5) that this method works very well in detecting moisture when used in a material such as sand, which does not contain hydrogen atoms. However, when used with a material like cellulose, which has a very high hydrogen content, this method does not work very well. There is also the effect of activating (making it radioactive) the sample due to the bombardment of the neutrons.

The alpha particle is absorbed in a very short distance, as shown

in Evans (4), which makes it impractical if the sample is very thick. For example, a sheet of paper will absorb most of the alpha particles incident upon it.

Beta particles are more penetrating than alpha particles. However, they are absorbed very easily, but, as shown by Dreshfield (6), they give good results for thin samples such as paper.

The gamma ray is similar to the x-ray, both being photons. However, their energy is higher and more penetrating. There is no restriction as to sample thickness with techniques using gamma rays. If more penetration is required, it may be accomplished by using higher gamma ray energy. Since gamma rays can be obtained by the decay of inexpensive radioisotopes, this method is relatively inexpensive. Gamma rays from a cobalt 60 source were used in this work. The reasons for this choice are as follows:

1. There is no major restriction on sample thickness.

2. The measurement of moisture will not affect normal drying processes.

3. Both types of moisture in hygroscopic materials will be measured.

4. The gamma ray beam can be collimated to a small area, thus making the moisture measurement almost a point determination.

5. The gamma ray source is low in cost.

Kazansky et al. (7) have used the gamma ray technique to investigate transient drying situations in quartz sand and a cellulose material. The source of gamma rays used was cobalt 60. The accuracy claimed by Kazansky in the determination of moisture contents was 0.5 percent in

quartz and 2.5 percent in cellulose, with the moisture content being based on the dry weight of each material. Since there was no discussion of counting rates as well as an inadequate description of the experimental equipment, there was no way of substantiating the indicated accuracy. The percentage error indicated by Kazansky appears to be too small; however, it is difficult to determine the statistical error (Appendix B) if the count rate is not given, even though the beam intensity was calibrated to moisture content as indicated in Kazansky's work. Without a description of the equipment there is no way of determining the counting technique. This leaves many questions unanswered about the discrimination level of the counter and the variation of the absorption of the gamma rays with moisture content. The statistical variation of the counting method and the variation of absorption rate of gamma rays with moisture content are the prime factors in determining the accuracy of the gamma ray method of determining moisture content.

Freeze-Drying

The preparation of dehydrated foods by the process of freeze-drying is now an established commerical process; however, the processing costs are still high when compared to more conventional dehydration techniques. These high costs are a result of slow drying rates, and the attempts to reduce the cost have concentrated on attempts to reduce the drying times. The work to reduce cost, however, must not destroy the advantage gained by the freeze-drying process, which is the superior quality of the product.

Koumoutsos and Sunderland (8) have said that the freeze-drying process has advantages over more conventional drying techniques for pre-

servation of the biological qualities. These advantages include better color, flavor, taste, and the reduction of product shrinkage. This reduction in shrinkage leaves the product with the property of being highly soluble, thus a freeze-dried food, upon immersion, will absorb water very quickly with this resulting in the restoration of its original appearance and synthesis.

The mechanism of the freeze-drying process has been examined by Harper (9). The ice phase recedes inward as the drying process proceeds and becomes surrounded by the dry layer of porous material. The heat that sublimates the ice to vapor must then be conducted through the dry layer to the phase front and the vapor must flow out through the same dry layer. The movement of vapor from the ice phase occurs by two processes: hydrodynamic flow as a result of a total pressure gradient and by diffusion resulting from a gradient in partial pressure of water vapor. Should the total pressure in the drying chamber be small in comparison to the vapor pressure of the ice in the product, the hydrodynamic flow pre-dominates and the diffusion flow is negligible. If the total pressure is high (near the triple point pressure), there is a negligible total pressure gradient and the vapor transport is by diffusion. Harper (9) has experimental results that indicate that diffusive transport predominates over hydrodynamic flow in the usual freeze-drying situation. Therefore, the water vapor in the vacuum chamber is important since the rate of diffusive transport depends on the difference in partial pressure of the water vapor at the ice surface and the vacuum chamber. If the partial pressure of the water vapor in the chamber is above the triple point pressure, thawing of the product would occur. In order to maintain

sufficiently low temperature for satisfactory product quality, the partial pressure of the water vapor must be kept well below the triple point pressure.

If a study of the mechanism of freeze-drying can result in a better understanding of the drying process, this understanding will lead to an improvement of the drying techniques and the reduction of the cost of dehydrated foods. Therefore, work done to establish the pressure of the vacuum chamber that results in the most rapid drying time will help reduce the cost. Also, determination of the thermal conductivity of dehydrated foods will result in a better understanding of the heat transfer problem involved in the freeze-drying process. A knowledge of two other transport properties is of interest. These two properties are permeability and diffusivity. Both will give a better understanding of the transport of vapor out of the dried porous material. Investigation of the receding of the phase front and observation of the moisture changes in the dry and wet material is also of interest. The interest is a result of the assumption being made about the phase front in attempts to obtain analytical solutions of freeze-drying situations. The assumption being made is that the phase front is a plane (with negligible thickness) as it passes through the material being freeze-dried. The observation of the phase front and moisture distribution is the concern of this work. This work will be conducted at different total vacuum chamber pressures to determine the effects of pressure on the phase front.

CHAPTER II

THEORY OF RADIATION ABSORPTION MOISTURE METER

The Interaction of Electromagnetic Radiation With Matter

Photons are classified according to their mode of origin, not their energy. Gamma rays are defined as electromagnetic radiation accompanying nuclear transitions. Unlike charged particles such as alpha and beta rays, a well-collimated beam of gamma rays shows a truly exponential absorption behavior in matter. The reason for this lies in the fact that photons are absorbed or scattered in a single event. That is, those collimated photons that penetrate the absorber have not interacted with the absorbing material, while the ones absorbed have been eliminated from the beam in a single event.

There are a number of processes that cause gamma rays to be scattered or absorbed. The three types of interaction that are of importance in the energy range from 0.1 to 6 Mev are the Compton effect, the photoelectric effect and the pair production effect. The other effects, which are minor, are presented by Evans (4). The Compton effect is one in which a photon is scattered by an electron of the atom in the absorbing material. The photon goes off in a different direction with decreased energy and the electron absorbs the remaining energy. Therefore, the collision is elastic and the direction of the photon is changed; also the energy of the photon is reduced. The photoelectric effect is the process in which a photon gives all its energy to a bound electron. This electron uses part of the energy to overcome its binding to the

atom and the rest as kinetic energy. The pair production is the process of absorption of photons in which a photon in the field of the nucleus produces an electron-positron pair, whose total kinetic energy is equal to the energy of the photon minus the mass energy of the two particles which have been created. This method can only take place when the energy of the photon is equal to or greater than the mass energy of the electron-positron pair. These three processes act independently of each other with one being more dominant at different energy levels or all three equally present, depending on the energy of the incident photon. Since cobalt 60 is the source of photons used in this work, and the absorbing material, meat is made up primarily of cellulose, the dominant effect involved in the absorption is the Compton effect with the other two effects being very minor.

The Relation of Attenuation to Moisture

The basis of all measurements of the absorption of gamma rays is based on the fact that the intensity of radiation decreases as it passes through material. The probability of a given photon passing through a distance x of the absorber without any interaction with the absorber is just the product of the probabilities of survival of the photon for each of the possible interactions. Therefore, the probability of the photon traversing the thickness x with a Compton collision is $e^{-\sigma x}$*, without a pair-production is e^{-Kx}, and likewise without a photoelectric effect is $e^{-\gamma x}$. Where σ is the linear attenuation coefficient for the Compton process, K is the linear attenuation coefficient for the pair-production effect, and γ is the linear attenuation coefficient for the photoelectric

*Symbols are defined in nomenclature section.

effect. Therefore, a collimated beam of gamma rays of intensity I_o will have a residual intensity

$$I = I_o e^{-\sigma x} e^{-\gamma x} e^{-Kx} \qquad (1)$$

of unaffected primary photons after it has traversed a thickness x of a particular material. Equation (1) may be reduced to

$$I = I_o e^{-(\sigma + \gamma + K)x}$$

or

$$I = I_o e^{-\mu_o x}$$

where μ_o is the total attenuation coefficient. This coefficient for gamma rays is a function only of the energy level of the incident photon and the absorber material.

For a given energy the mass attenuation coefficient can be expressed as the following:

$$\frac{\mu_o}{\rho} = C N_a Z$$

where Z is the atomic number, N_a is Avogadro's number, ρ is the density of the absorber, and C is the constant of proportionality.

It is seen that a given material will have a constant mass absorption coefficient if the energy of the incident radiation is constant. If a collimated beam is incident upon a material in which the density is changing, then the change in the residual intensity is a direct function of the

change in density of the absorber. Therefore, when used with meat to study the freeze-drying process, the only property that is changing is the density. The change of density is caused only by a change in the moisture content. Therefore, if a collimated beam of gamma rays is passed through the same point in the meat during the drying process, the change in the attenuation of the gamma rays through the meat can be related directly to the change in moisture content of the meat.

Since gamma ray absorption is exponential, the equation of absorption can be written for a sample between its wet and dry state. The equation is

$$I_w = I_{dr}\, e^{-ku} \tag{2}$$

where I_w is the residue intensity of a gamma ray beam after passing through a wet sample and I_{dr} is the residue intensity of the same gamma ray beam passing through the same dry sample; also k is a constant that is determined experimentally and u is the moisture content. The constant k incorporates x, ρ_{dr} (density of dry product), Z, A, N_a, $_e\mu$ and all the constants involved in the exponent of Equation (1). Therefore, the calibration of the gamma ray beam to determine moisture content directly during drying processes at any moment of time can be obtained and the moisture content is given by the equation

$$u = \frac{1}{k} \ln \frac{I_{dr}}{I_w} \tag{3}$$

Since the current work is concerned with the application of a gamma ray moisture meter to the process of freeze-drying, there is no need to determine the constant k or I_{dr}, because the ice passes to a vapor and there is no intermediate moisture content. The sample is either wet or dry. Therefore, if a one-dimensional drying situation can be obtained for the freeze-drying process, the gamma ray technique can be used to determine the rate at which the ice phase recedes from the exposed face of the sample.

CHAPTER III

EXPERIMENTAL INVESTIGATION

Instrumentation and Equipment

Radiation Beam

In order to generate the gamma ray beam, the radioisotope Co^{60} was placed in a lead container (see Figure 1) and a 1/16 inch diameter hole eight and one half inches long was drilled through the lead to collimate the radiation emitted from the cobalt. The lead container was placed as near the meat sample as possible in order that the radiation beam be very small when it passed through the sample. The length of the hole through the lead container also determined the degree of collimation obtained, since radiation is emitted in a diffused manner from the cobalt. The degree of collimation obtained at the meat sample was calculated (as shown in Appendix A) and also determined experimentally by exposing a strip of photographic film to the beam and measuring the exposed area on the film. The 1/16 inch diameter hole eight and one half inches long was found to collimate the beam sufficiently at the section where the meat sample was being placed, with the collimated beam having approximately 3/16 inch diameter area at the meat sample.

To conform with safety standards when using radioisotopes, there was a minimum of seven inches of lead placed around the radioactive source in all directions. This lead allowed the area to be considered a safe working area. Since the gamma ray beam emitted from the small

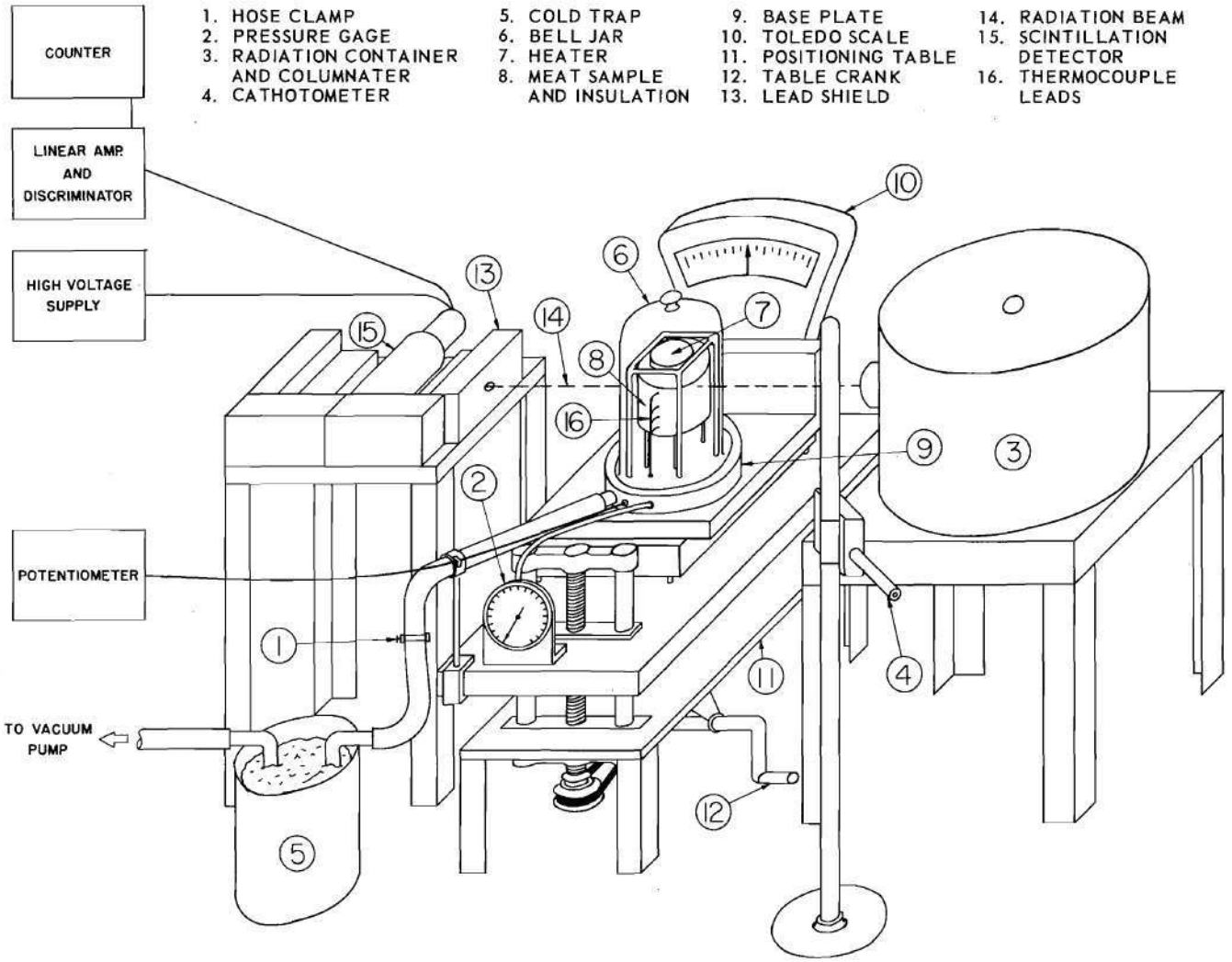

Figure 1. Experimental Set-up.

hole was of very high intensity and would cause damage if there was prolonged exposure, there was provided a means by which the cobalt source could be lowered away from the hole in the lead container to minimize the radiation emitted from the hole during set-up of the meat sample. This positioning mechanism consisted of a rod attached to the cobalt container with a screw to allow for the positioning.

Scintillation Counting

The intensity of the radiation beam after it had passed through the meat sample was determined by use of a scintillation counting technique. This technique consisted of placing a crystal in the radiation beam and using a photomultiplier tube to measure the amount of light that was produced by the ionizing radiation as it was absorbed by the crystal. The pulses from the photomultiplier tube were then counted and the number of counts was proportional to the intensity of the gamma ray beam. The scintillation detector used in this work was a Harshaw NaI(Tl) Scintillation Detector, Type 12512, with a crystal three inches long and three inches in diameter.

The height of the electronic pulse output of the photo tube was proportional to the incident gamma ray energy. Thus, to eliminate all the background radiation and the photons that had collided with the water molecules, but continued in the same direction, to reach the detector crystal, a Franklin Electronics Linear Amplifier and Discriminator was placed in the system after the linear amplifier of the photomultiplier tube. This discriminator would eliminate all electrical pulses of the photo tube that were lower or higher than a predetermined range. Therefore, as previously mentioned in Chapter II, the scatter

radiation (radiation that has collided with a water molecule) has a lower energy than the original photon; thus, by setting the discrimination level to count only the two maximum peaks of cobalt energy (1.17 and 1.33 Mev), all other energy ranges of lower or higher instant photons were eliminated. The output of the discriminator was counted by use of a Systron Counter, Model 1032.

In order to provide an accurate method for determining the time of each counting period, a pulser was made that had an output of sixty pulses a second. The pulses were counted by a Tracerlab Versa/Matic II Scaler on which a pre-set count was set. The starting and stopping of the Tracerlab counter triggered the counting circuit of the Systron counter. This method was found to be accurate to within one ten-thousandth of a second.

The above timing and counting technique was used for the first two tests; however, for the third test, a Tracerlab Scaler-Spectrometer Counter was used. This instrument had a timing circuit that was as accurate as the pulse counter method used in the first two tests.

Mechanical Equipment

The vacuum chamber consisted of a bell jar eight and one half inches in diameter and fifteen inches high, which was placed on a base plate and sealed with a rubber gasket. A vacuum pump* was connected to the base plate by a rubber hose in order to place a constant tare on the scale (Figure 1). A cold trap was placed between the vacuum pump and the vacuum chamber and connected with rubber hose. A mixture of acetone

*Welch Scientific Corp. Model 1402, with a capacity of 0.35 cubic feet per minute at the total chamber pressures used in this test.

and dry ice was used in the cold trap (see Figure 1).

The bell jar and base plate were placed on a Counter-Balanced Toledo scale, which could be read to 0.01 lbs., with the scale being placed on a positioning table. The positioning table consisted of a pulley and crank that operated two jack screws, thus allowing the positioning of the table surface.

The determination of the depth at which the beam was passing through the meat sample was accomplished by placing a scale on the jar and sighting on the top of the meat sample with a cathotometer, then observing the different levels on the scale as the table positioned the sample.

The heater used in the vacuum chamber was the same diameter (three inches) as the meat sample in order to minimize the addition of heat to the side of the sample. The heater was controlled by a powerstat.

Temperature Measurement

The temperature measurements made in this work were taken at different levels of the meat sample during the drying process. These temperatures were measured with copper-constantan thermocouples inserted in the meat sample at the different sections of temperature measurement. The thermocouples were made from No. 30 copper-constantan wire and the two wires were attached by a heat welding process, with there being no other metal involved in the junction of the two wires. The junction formed at the end of the wire and the wire only was inserted into the meat at the proper section by drilling a very small hole in the frozen meat sample. It was observed that during the drying process there was no drying around the thermocouples. However, it was found that if

hypodermic needles were used to insert the thermocouple, with the thermocouple wire inside the hypodermic needle, there was a large amount of drying around the thermocouple junction in the frozen meat due to heat conduction into the frozen meat by the hypodermic needle.

The thermocouple wire was passed through the base plate by Thermo Electric vacuum thermocouple feed-throughs. The thermocouple voltage output was determined by a Leeds & Northrup Potentiometer, Model 8662.

Pressure Measurement

The measurement of absolute pressure in the vacuum chamber was accomplished by the use of a Wallace-Tiernan Absolute Pressure Gage, calibrated from 0.2 to 20.0 millimeters of mercury. The pressure gage was connected to the base plate by a 1/4 inch inside diameter flexible teflon tubing, which was 26 inches from gage to base plate. This flex hose placed a constant tare on the scale.

Experimental Procedure

Sample Preparation

The meat sample used in this work was obtained from a "top of the round" cut of utility beef. The samples were carefully cut from frozen meat to insure a uniform grain direction from top to bottom and to insure essentially one dimensional drying. The direction of the fibers and fatty inclusions have a marked effect on the rate and direction of drying. However, by using the previously mentioned cut of meat, it was possible to obtain a sample two inches thick and three inches in diameter that contained very little fatty inclusions and also very straight grain direction.

An attempt to wrap the sample in aluminum foil to shield the

sample from side drying resulted in a considerable amount of side drying and this method was abandoned. However, it was found that by wrapping the sample in plastic wrap and tape there was no side drying of the sample.

After the sample was wrapped, the thermocouple holes were drilled and the thermocouples inserted. Tape was then placed over the thermocouple holes to prevent any vapor from escaping through the holes in the plastic (Figure 2). The sample was surrounded by two inches of fiber glass insulation with aluminum foil on the outside of the insulation to retard heat being radiated to the insulation (Figure 3). The sample was placed into the vacuum chamber with the heater placed over the exposed face of the sample. The distance between the heater and sample was one quarter of an inch.

Drying of the Sample

The experimental procedure used to dry the sample and observe the progress of the phase front in the meat sample with the gamma ray beam is as follows:

1. The power to the high voltage supply, discriminator and counter was turned on and a minimum warm-up time of three hours was provided.

2. The cold trap was filled with acetone and dry ice.

3. The sample was prepared as mentioned in the previous section.

4. The meat sample, insulation and thermocouples were weighed before they were placed in the vacuum chamber.

5. The cathotometer was set up and set on the radiation beam hole.

6. The scale and its supporting table were leveled.

7. The sample was placed on a support stand on the bell jar base plate.

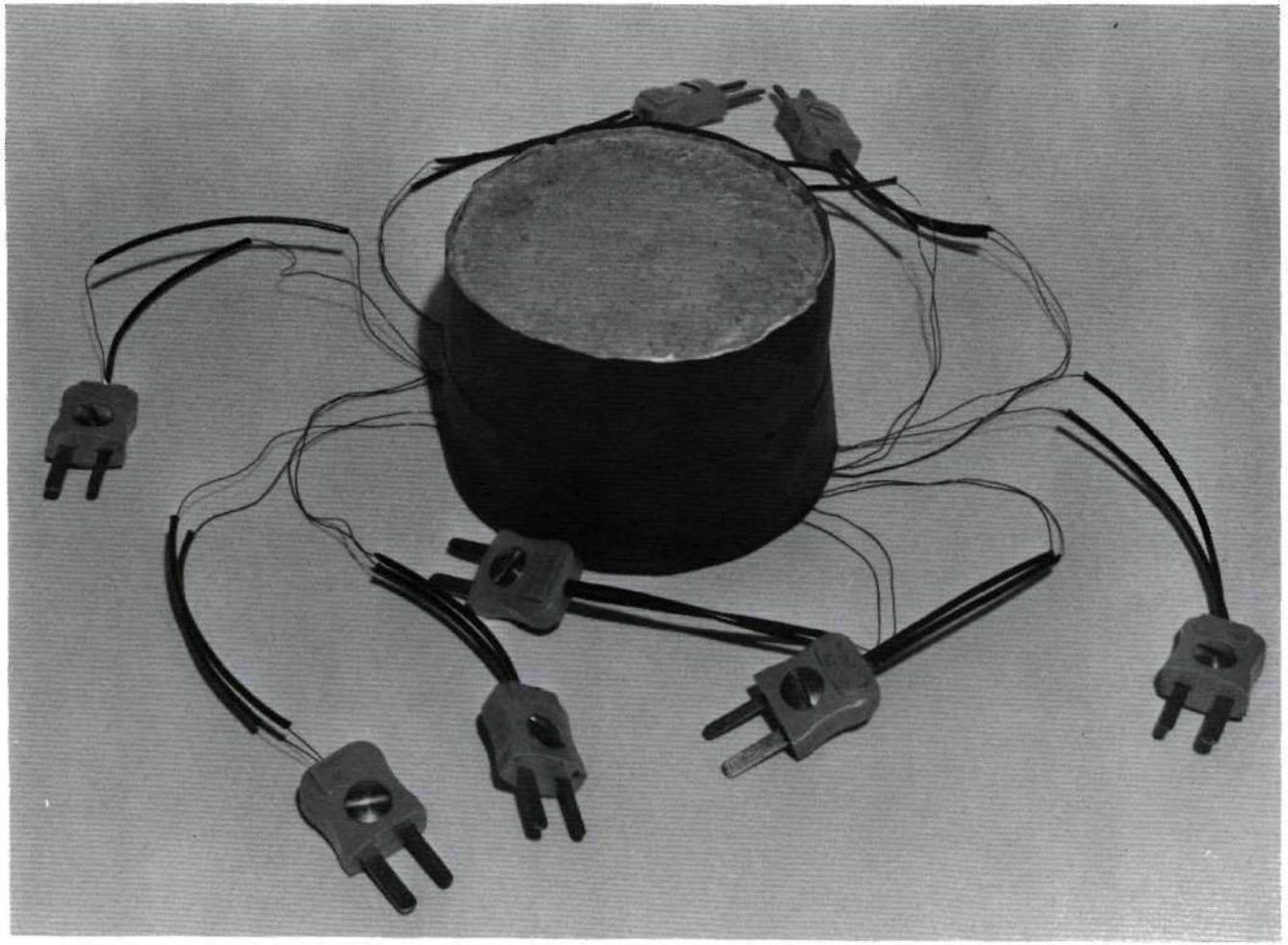

Figure 2. Meat Sample and Vapor Seal.

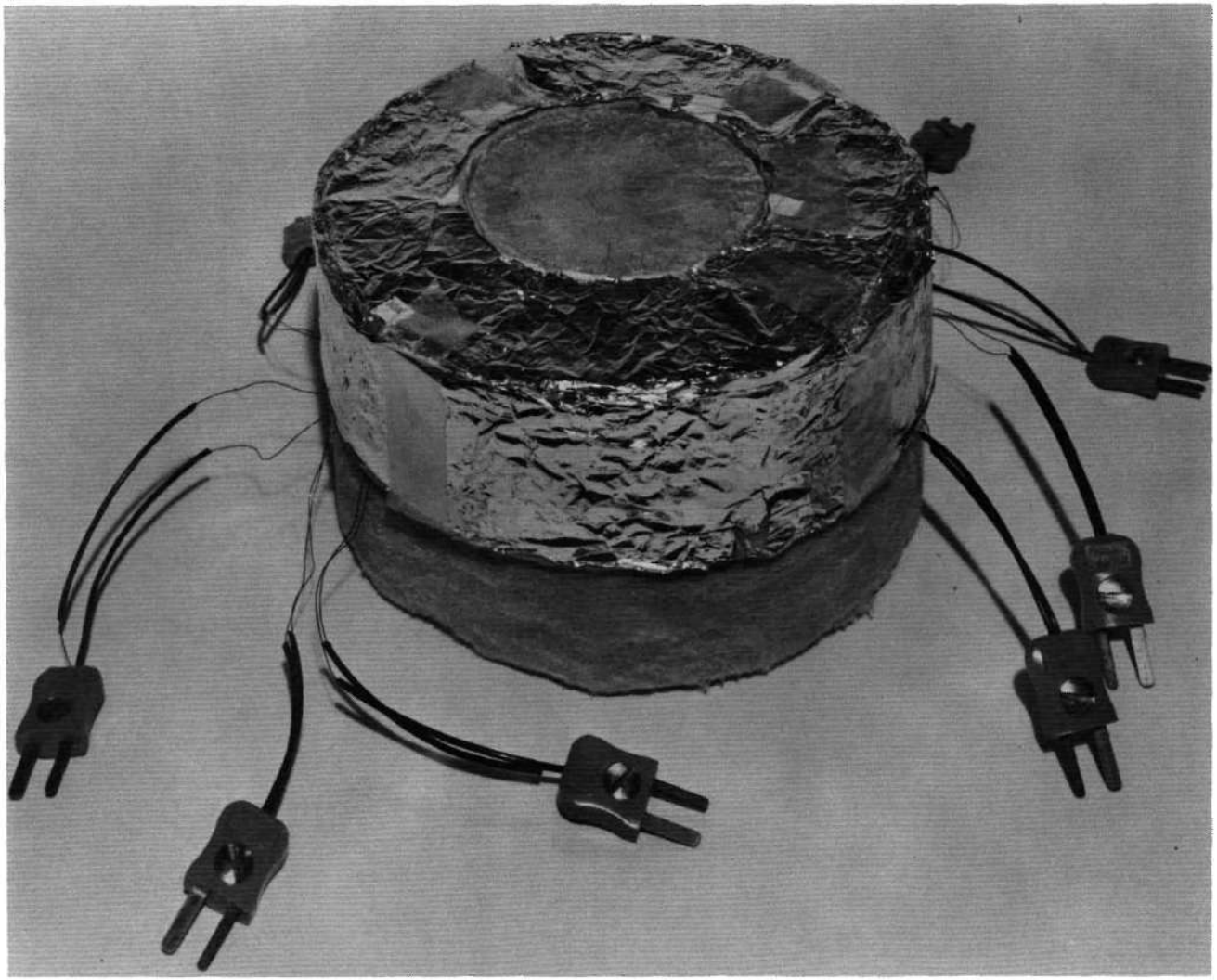

Figure 3. Meat Sample and Insulation.

8. The thermocouples were connected and the thermocouple used to control the surface temperature of the sample was placed as close to the surface of the meat as possible by using a small wire to make a hole in the frozen meat just under the surface.

9. The heater was placed over the sample and connected to the various controls.

10. The bottom of the bell jar was coated with vacuum grease and placed on the base plate with the rubber gasket as a seal.

11. The vacuum pump was started.

12. The level of the table was adjusted until the top of the meat was in line with the center of the radiation beam. This was accomplished by the cathotometer.

13. A scale graduated in 1/32 of an inch was taped to the bell jar; thus, allowing the table to be moved to position the radiation beam at any level in the meat and the distance from the surface of the sample could be determined accurately by use of the graduated scale and cathotometer.

14. The radiation beam was placed at 1/8 inch levels from the free surface and a pre-set counting time of 100 seconds was used in test three and a pre-set time of 166.67 seconds was used in tests one and two.

15. The radiation source was adjusted in the container so that a count rate of approximately one million counts a minute could be obtained through the frozen meat sample.

16. Counting was then started at 1/8 inch intervals, starting at 3/16 from the exposed surface of the sample and the time recorded at the end of each count. The table was then moved so that the beam passed

through the sample 1/8 inch lower and the counting started again. This process was carried out continuously throughout the test.

17. The power to the heater was turned on.

18. Temperature readings were taken with the potentiometer every half hour throughout the sample and the surface temperature checked every ten minutes and this surface temperature was held at 100° F. throughout the entire test.

19. The pressure in the chamber was held constant through the test by placing a hose clamp on the rubber hose between the vacuum chamber and the cold trap. Thus, by tightening or loosening the clamp, the pressure in the vacuum chamber was controlled.

20. The initial weight reading on the Toledo scales was taken with the table positioned so the radiation beam was passing through 1/8 inch and 1 inch from the free surface. Thereafter they were taken when the table was set at these positions and the time was recorded when each reading was taken.

21. The gamma ray beam was exposed to an aluminum block that had approximately the same attenuation as a half dry sample of meat, to check the electronic equipment for drift in the counting rate or changes in the discrimination level. This was done each time a complete traverse of the sample by the gamma beam had occurred.

22. The test was completed when the residue beam intensity indicated the sample was completely dry at 1 inch from the free surface.

23. The Toledo scales were calibrated during each test by placing gram weight on the scale and observing the reading.

24. The pump was turned off and the meat sample insulation and

thermocouples were removed and weighed to check total weight loss.

The above procedure was followed to measure the transient phase front position, temperature distribution and weight loss of the sample at chamber pressures of one, two, and three torr.

CHAPTER IV

PRESENTATION AND DISCUSSION OF RESULTS

The curves obtained for the receding of the ice front by use of the gamma beam are shown for the three different pressures considered in Figures 4 through 6. There is no indication of moisture content on the vertical scale of the beam data, due to the peculiarity of the drying process. The assumption that the ice front is a plane was checked by the successive drying of samples and cutting them apart to check the ice front. By visual inspection, it was found that the ice front may be assumed to be a plane. Thus, there is a step change in moisture content of the meat and for this reason the radiation beam is exposed either to wet frozen or dry meat with no intermediate stage between the two moisture contents. Since the beam is 3/16 inch in diameter (Appendix B) at the meat sample, a situation exists in which the phase front is in the center of the radiation beam. Thus, from the calibration curves, the radiation data would indicate that the sample was half wet and half dry at this particular position. This would not be the case, due to the area of the radiation beam. This would only indicate that the phase front was in the center of the circle formed by the radiation beam. Therefore, when the beam intensity curves of Figures 4 through 6 are half way between the wet and dry ends of the curve, the phase front was just passing that particular position. Thus, for this reason, moisture content cannot be placed on the vertical scale of these figures, only wet where the curves have not started to rise and dry when the count rate rise is complete.

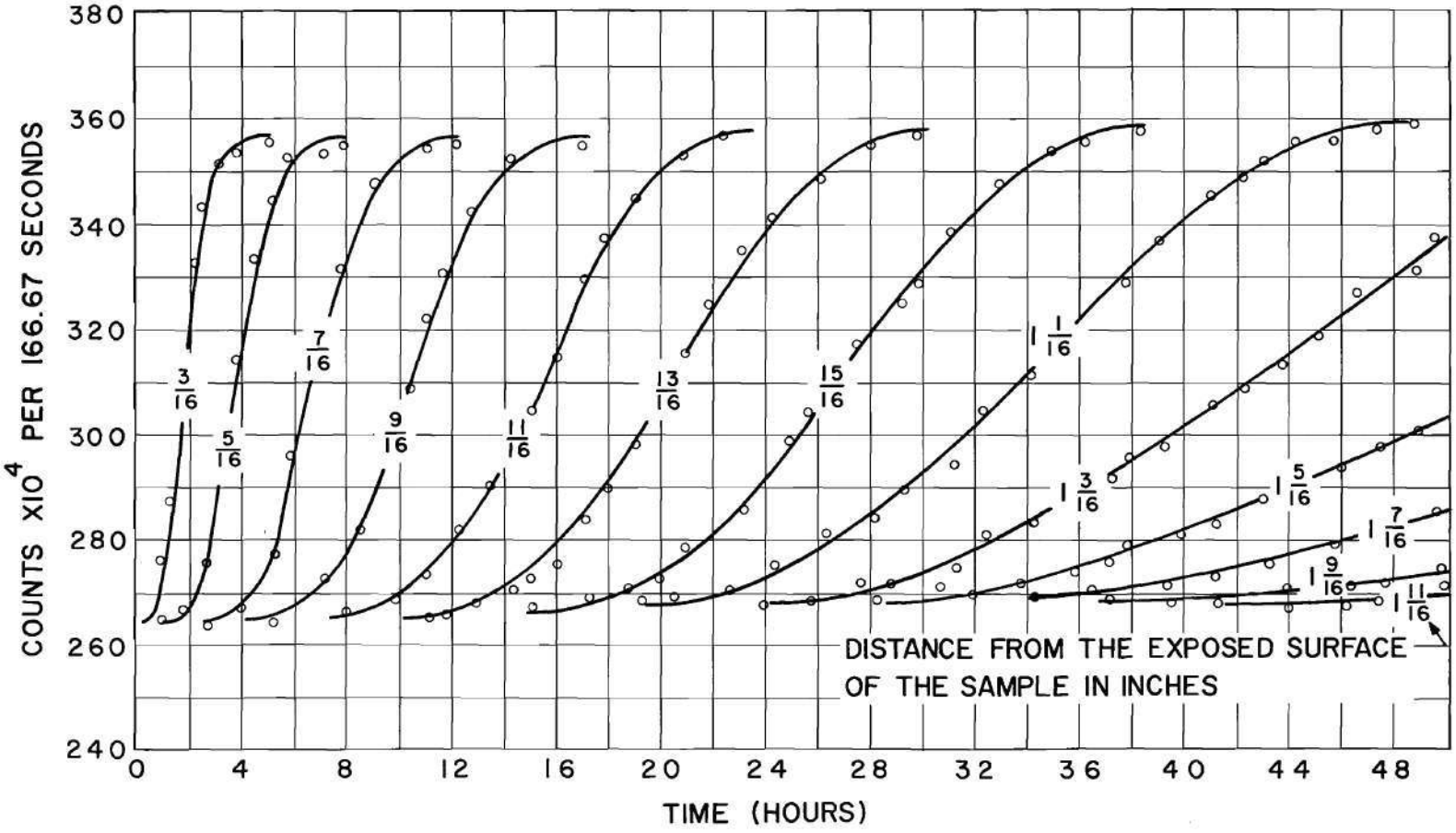

Figure 4. Counts Versus Time at Positions from the Exposed Sample Surface (p = 1 Torr).

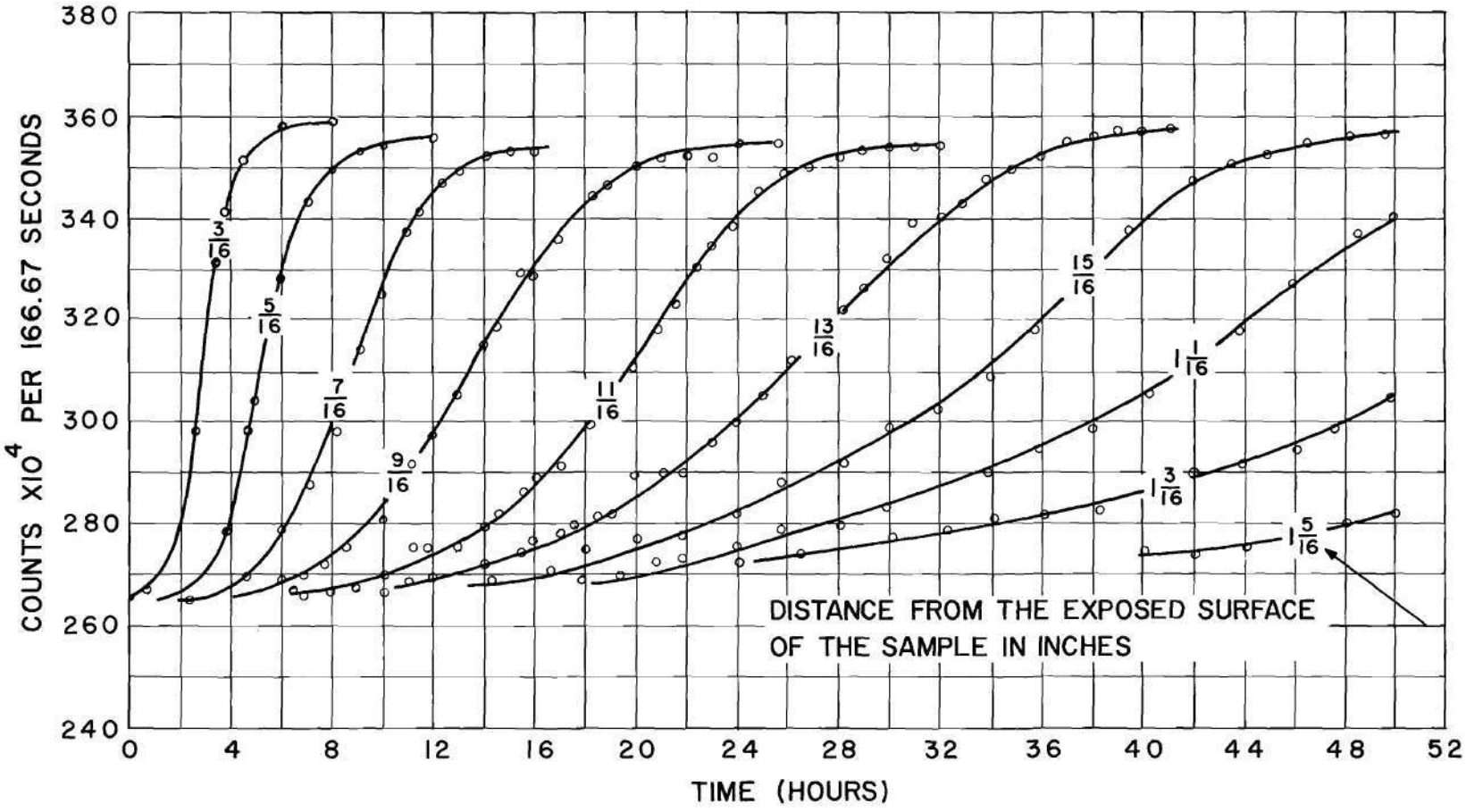

Figure 5. Counts Versus Time at Positions from the Exposed Sample Surface (p = 2 Torr).

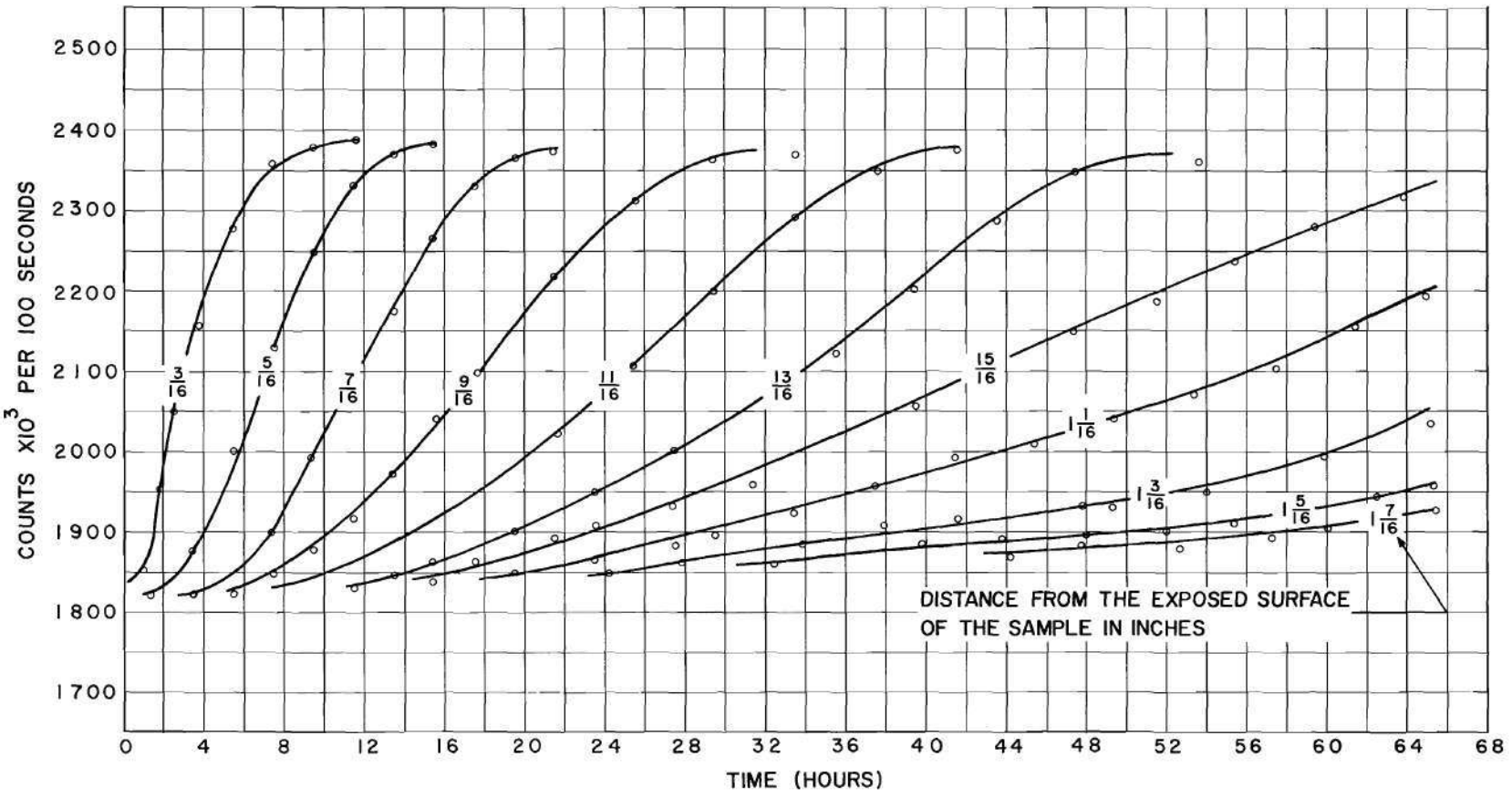

Figure 6. Counts Versus Time at Positions from the Exposed Sample Surface (p = 3 Torr).

Also, the mid-point of the beam intensity rise indicates that the phase front is at this particular position. Therefore, when the center of the radiation beam is set at each position from the free surface, the radiation intensity may start to rise before the phase front reaches that position because of the zone of effect that the radiation beam has. For example, in Figure 4 at 5/16 of an inch from the free surface, the radiation intensity indicates the position starting to dry at approximately one hour after the start of the test. This indicates that the phase front has passed into the zone of effect of the radiation beam when the curve starts to rise and is at the 5/16 inch position when the curve is half way up the scale and completely out of the zone when the curve reaches its maximum. Thus, when the curves are half way up the scale, the phase front is at the distance from the free surface indicated on each curve.

Data was taken at each position after the curve had reached a maximum and there was found to be no change in the moisture content. Thus, when the curves reached a maximum they became straight lines of zero slope with no change through the test, which indicates there was no change in the moisture content in the dry meat. Similarly, data was taken in the wet frozen section of the meat and it was found that the curves indicated no change in moisture content; however, there was a slight rise in the wet frozen section curves before the arrival of the phase front that may be explained by side drying. It was found at the end of each test that there was a small amount of side drying (approximately 1/16 of an inch at a drying depth of one inch), which will explain a slight rise in the wet portion of the curves before the phase front

reached that position.

Based on the knowledge of the beam geometry and the phase front shape, it may be concluded that when the phase front is at a particular distance from the free surface, the mid-point of the beam curve rise indicates the phase front is at that level.

The curves obtained for the temperature data for the different pressures considered are presented in Figures 7 through 9. It can be observed that the temperature at each position reached a minimum just before it started to rise. It may also be observed that the temperature in the frozen section of the meat is a function of the chamber pressure; the lower the pressure the lower the temperature of the frozen meat juice. Also, from the temperature data it may be observed that the minimum temperature in the sample occurs at or near the phase front.

The weight loss for each test (1, 2 and 3 torr chamber pressure) is presented in Figure 10 and the samples weight loss checks with those weights taken before and after each test. It can be seen from these data that the chamber pressure of one torr results in a faster drying rate with the two and three millimeters drying about the same. The temperature data also verify this observation. However, the beam data indicate that one torr chamber pressure resulted in a faster drying time with the three millimeter test being much slower than the two millimeter test. The reason for this discrepancy in the three millimeter test lies in the fact that there was a large amount of distortion of the meat sample. This distortion of the sample resulted in a change of the ice front rather than a change in the ice front due to normal freeze-drying. It was observed at the end of the test that the sample had shrunk in dia-

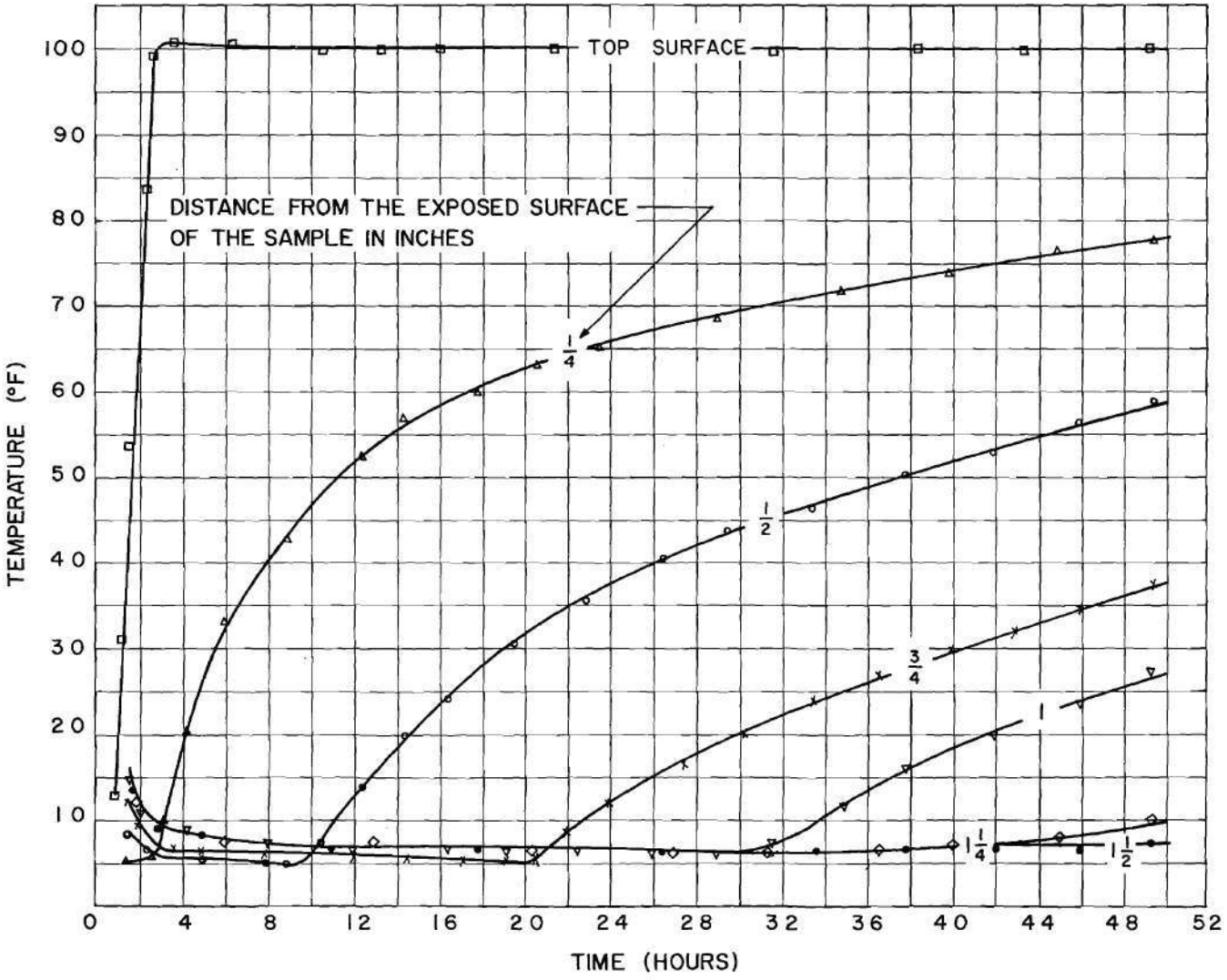

Figure 7. Temperature Versus Time at Positions from the Exposed Sample Surface (p = 1 Torr).

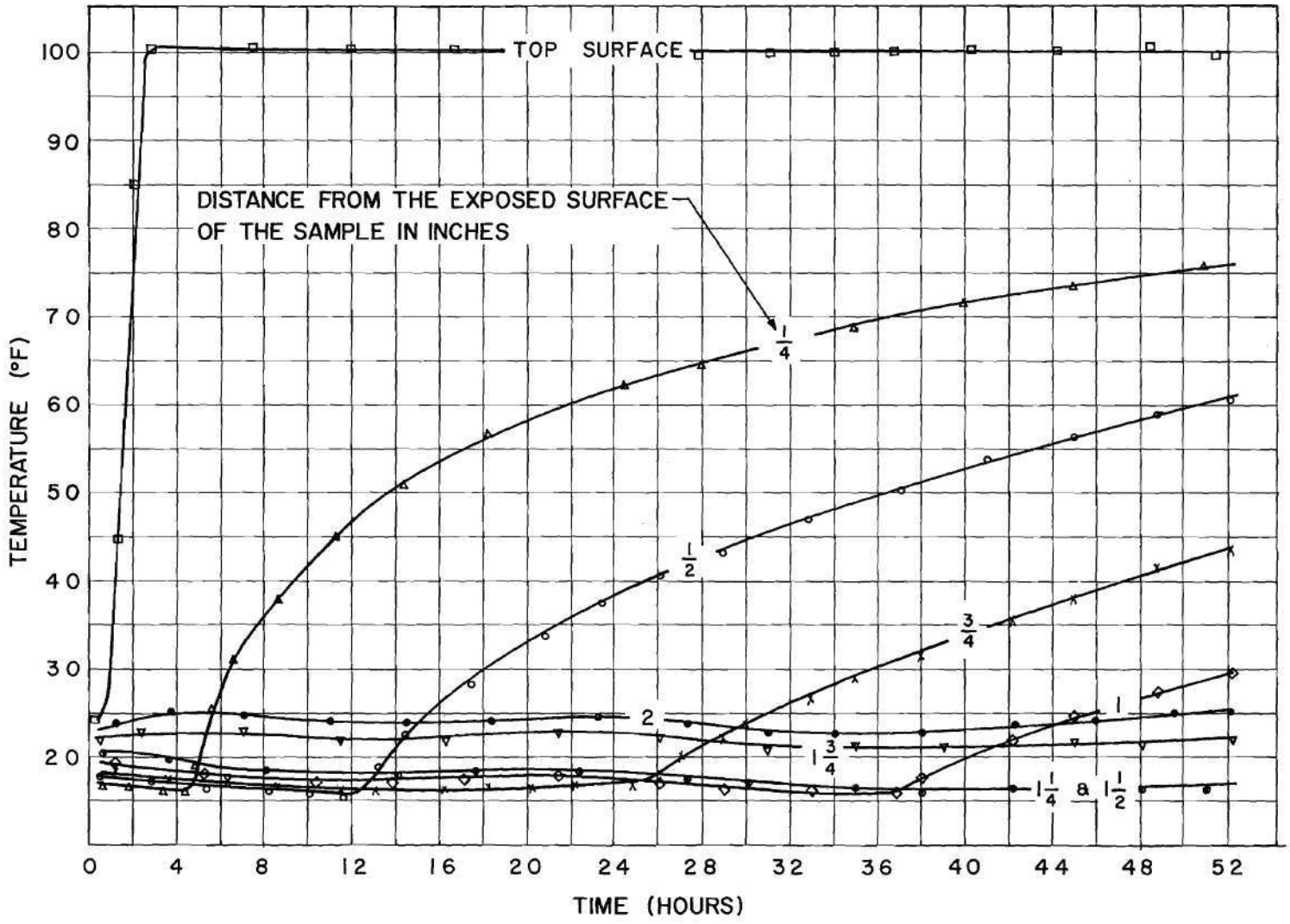

Figure 8. Temperature Versus Time at Positions from the Exposed Sample Surface (p = 2 Torr).

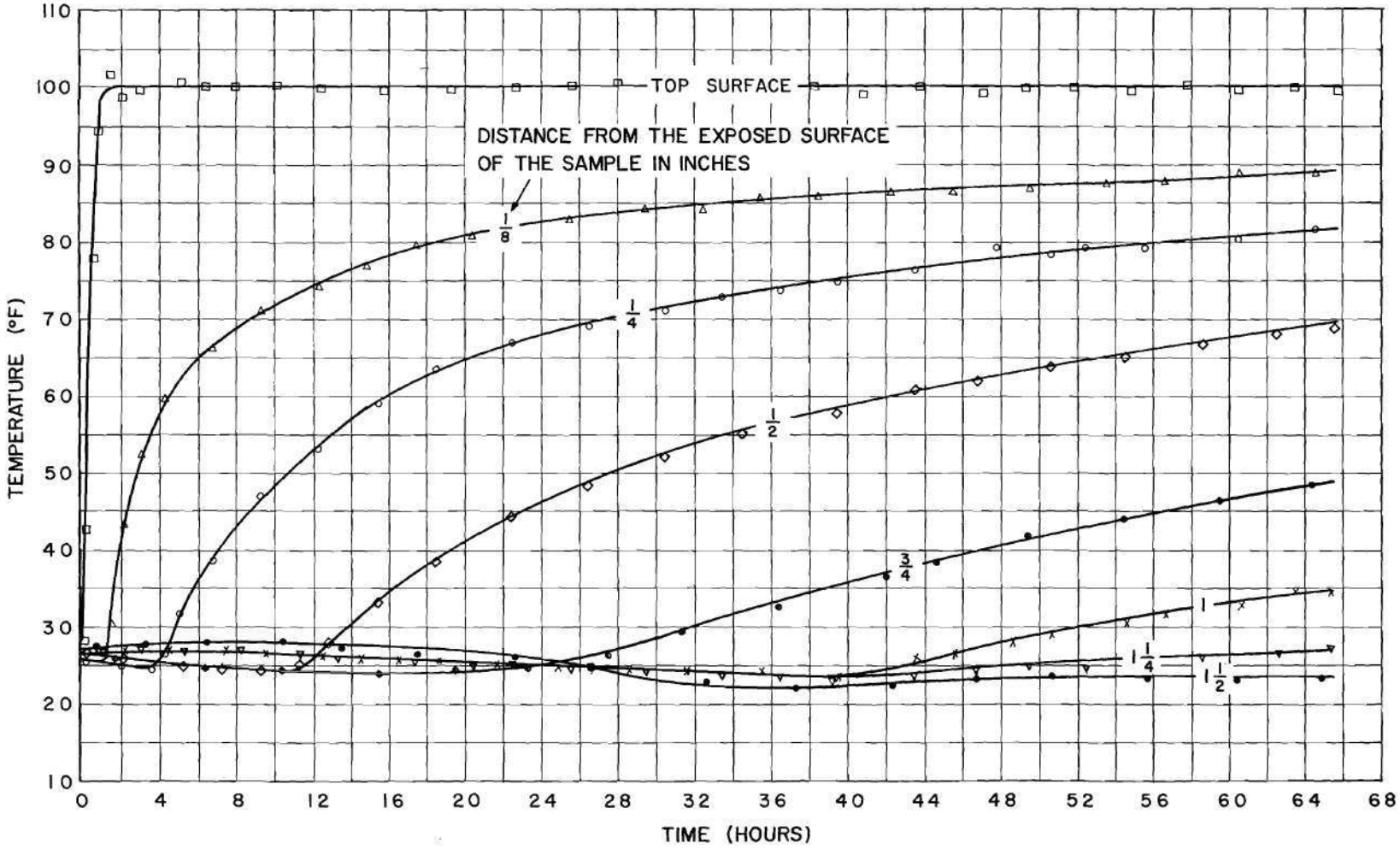

Figure 9. Temperature Versus Time at Positions from the Exposed Sample Surface (p = 3 Torr).

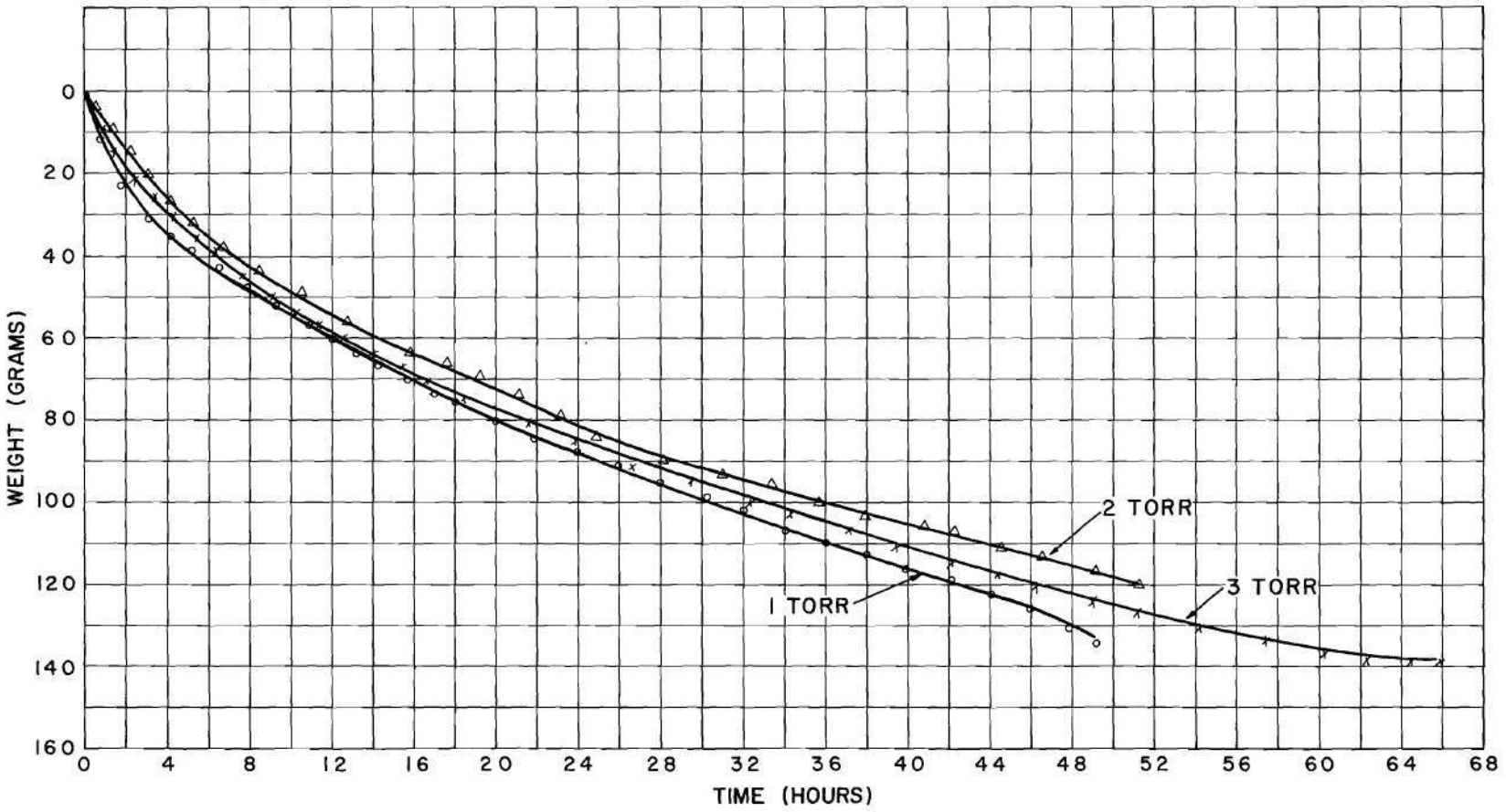

Figure 10. Weight Loss of Sample Versus Time.

meter by about 1/4 inch and increased in thickness by about 1/8 of an inch. This deformation made the interpretation of the beam radiation data difficult. The distortion of the sample may be a result of the high chamber pressure, because a second test was attempted at three millimeters of mercury and the same result was obtained. Also, another reason for this conclusion about distortion of the sample with high chamber pressures lies in the fact that very little distortion was obtained at two millimeters and none at all at one millimeter of mercury chamber pressure.

A cross plot of the temperature and count rate distribution for the three chamber pressures considered is presented in Figures 11 through 16. The data indicate that the temperature minimum occurred at about the same position as the ice phase front. Again these data bear out the fact that the three millimeter test results do not correlate. The assumption that the ice front was at the minimum temperature has been made in references 8 and 9 and is substantiated by the data obtained in this work.

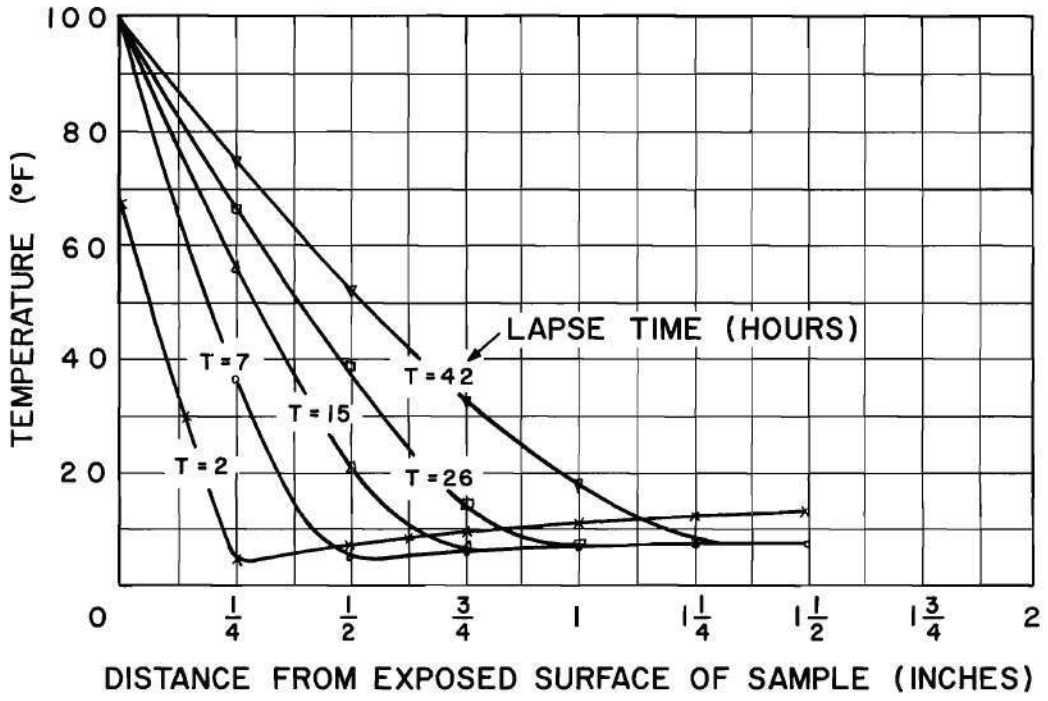

Figure 11. Temperature Versus Position (p = 1 Torr).

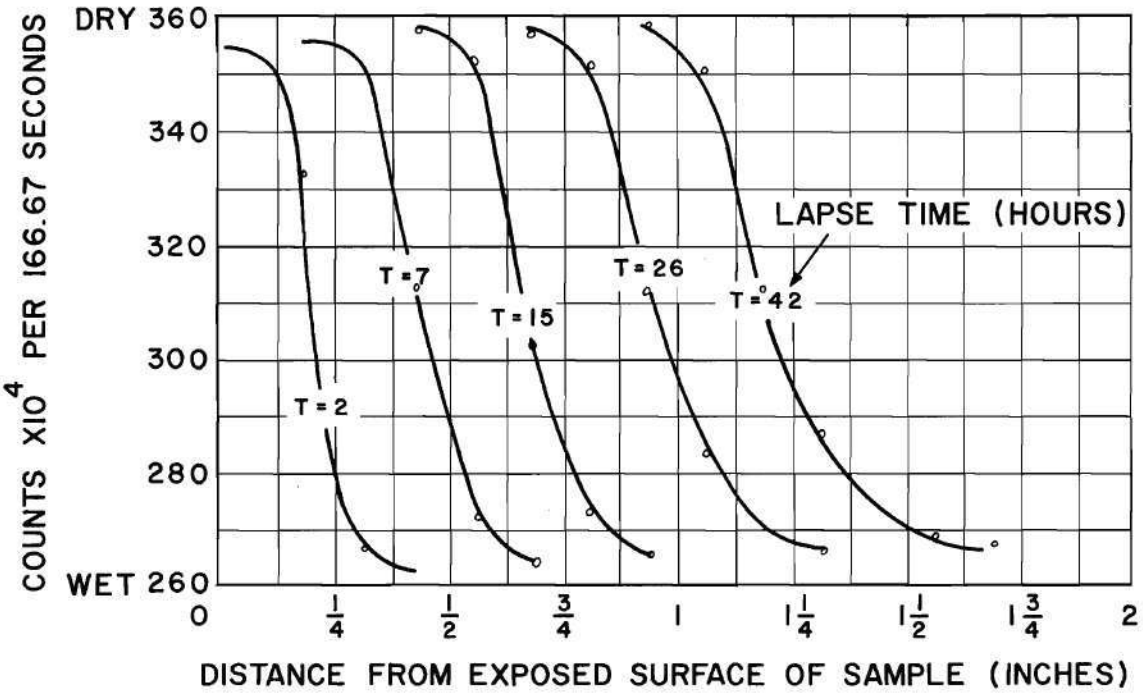

Figure 12. Counts Versus Position (p = 1 Torr).

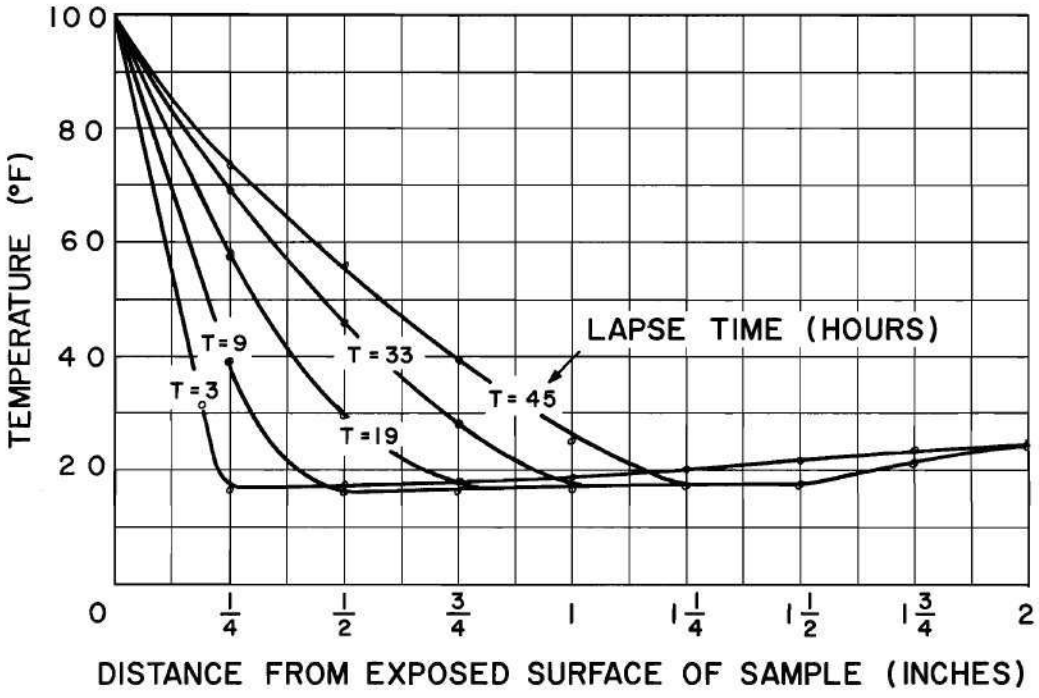

Figure 13. Temperature Versus Position (p = 2 Torr).

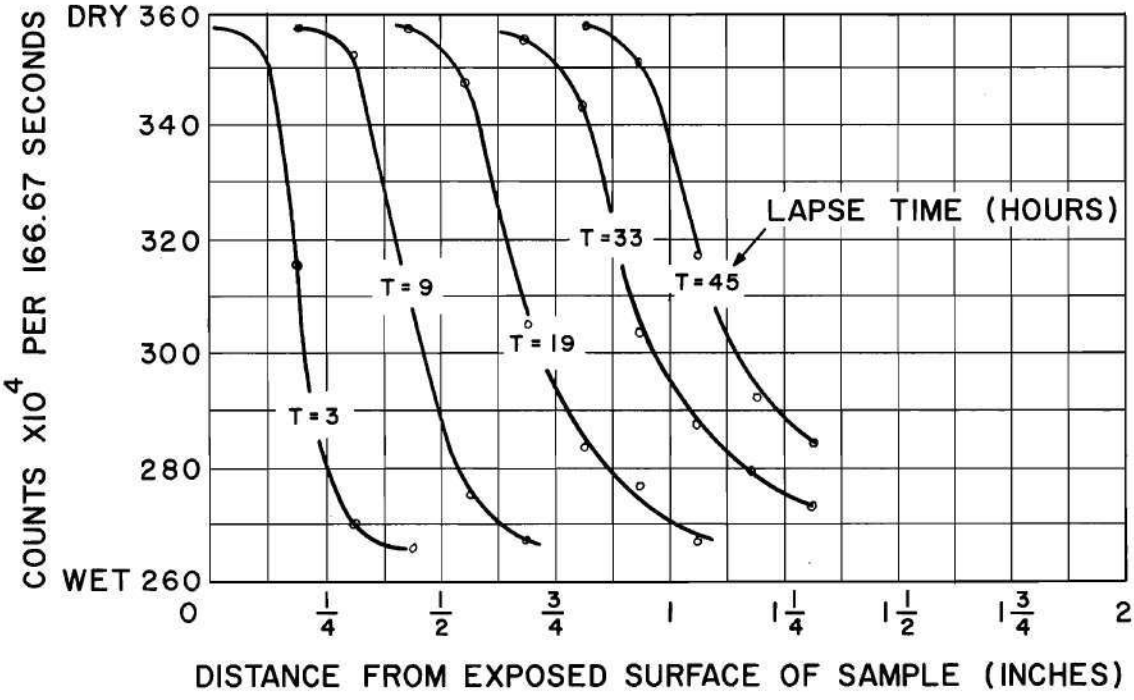

Figure 14. Counts Versus Position (p = 2 Torr).

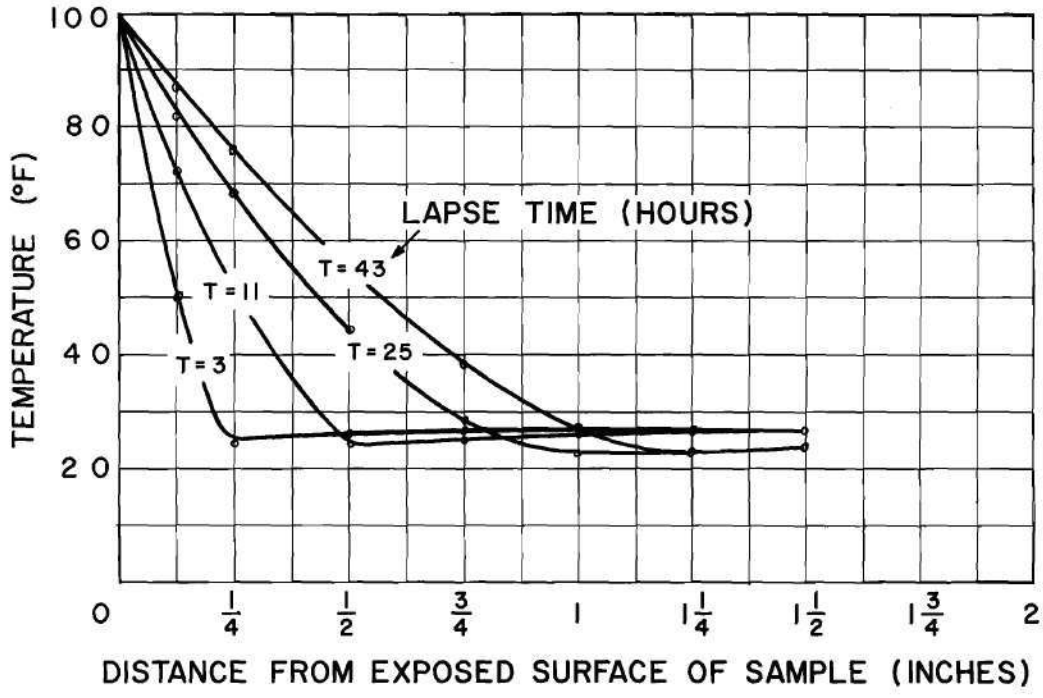

Figure 15. Temperature Versus Position (p = 3 Torr).

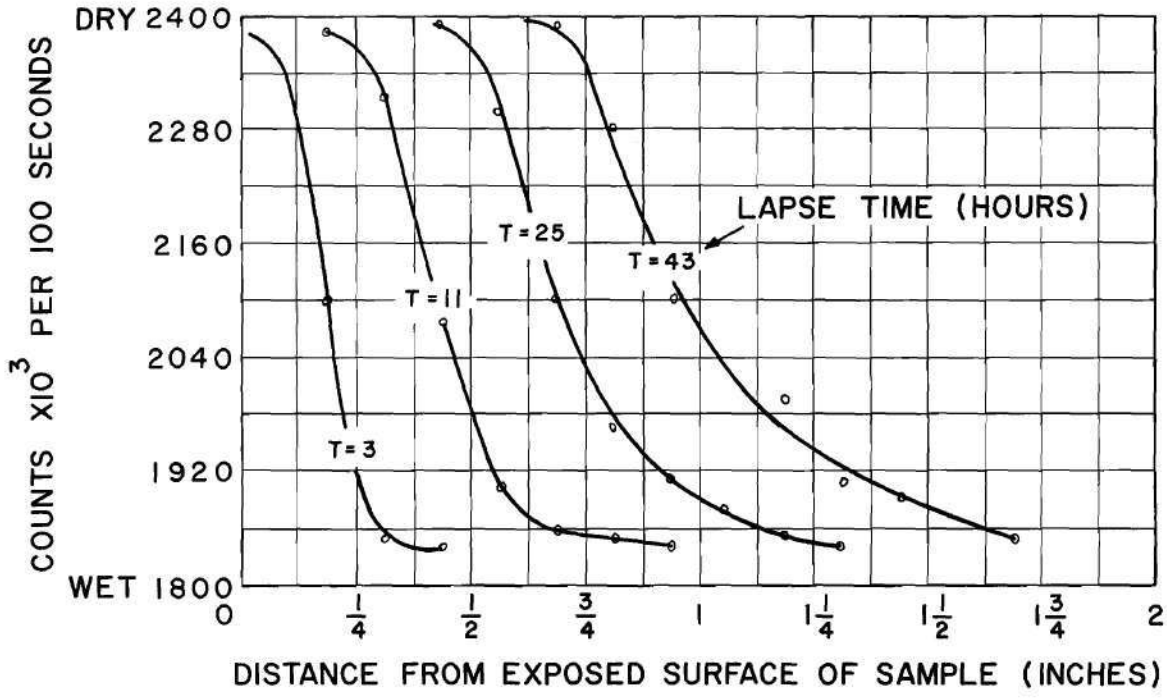

Figure 16. Counts Versus Position (p = 3 Torr).

CHAPTER V

CONCLUSIONS AND RECOMMENDATIONS

An experimental investigation has been made to determine the local moisture content during freeze-drying of meat at a total chamber pressure of one, two, and three torr.

The conclusions drawn from this investigation are:

1. The attenuation of gamma rays will give good results in the determination of moisture content.

2. There was no more than a three percent variation in the vapor density in the dry layer of the meat during the freeze-drying.

3. Drying of the meat takes place at a plane phase front and appears not to take place over a volume in the meat.

4. There was faster drying obtained at one torr chamber pressure than at two or three torr.

5. There is little change in the temperature of the frozen meat until the phase front passes.

6. The radiation beam could be used to indicate when the sample is completely dry by passing the beam through the meat sample from the free surface to the bottom of the sample.

The following items are recommended as a logical extension of the work which has been presented in this thesis:

1. The gamma ray beam technique should be applied to a drying situation which would allow the calibration of the residue intensity to local moisture content.

2. Work should be done to reduce the beam diameter, thus measuring the moisture content over a much smaller area of the sample.

3. An investigation should be conducted with lower or higher energy gamma rays to determine if better results are obtained when the incident photon is of a different energy than the gamma rays obtained from the radioisotope cobalt 60.

4. Instrumentation should be obtained to handle higher gamma ray flux which would allow the technique to be applied to faster drying processes.

APPENDIX A

BEAM GEOMETRY

Radiation is emitted from a radioisotope in a diffused manner. To form a collimated beam of gamma-rays, a long barrel of lead must be used as shown in Figure 17.

A reduction in the diameter of the radiation beam at the sample results in a more localized moisture measurement. The diameter of the beam may be reduced by increasing the barrel length and by making the hole through the lead smaller in diameter. Difficulties arise with both methods. If more lead is used to increase the length of the barrel, it becomes difficult to drill a small straight hole through a long section of lead. (The hole must be straight or no radiation will be emitted through it.) Also, a reduction in the hole diameter will also generate drilling problems. Another problem with efforts to decrease beam diameter by these methods lies in the reduction of radiation flux through the hole. Therefore, if a high count rate is required, the intensity of the source must be increased.

The beam geometry used in this work is shown in Figure 17. The diameter of the beam as it passed through the meat sample was less than 3/16 of an inch in diameter. Thus, making the zone of moisture measurement in the meat no larger than 3/16 of an inch in diameter. The geometrical results of Figure 17 were checked by exposing a strip of photographic film to the radiation beam at the scintillation detector and measuring the diameter of the exposed area of the film. This technique verified the geometrical results.

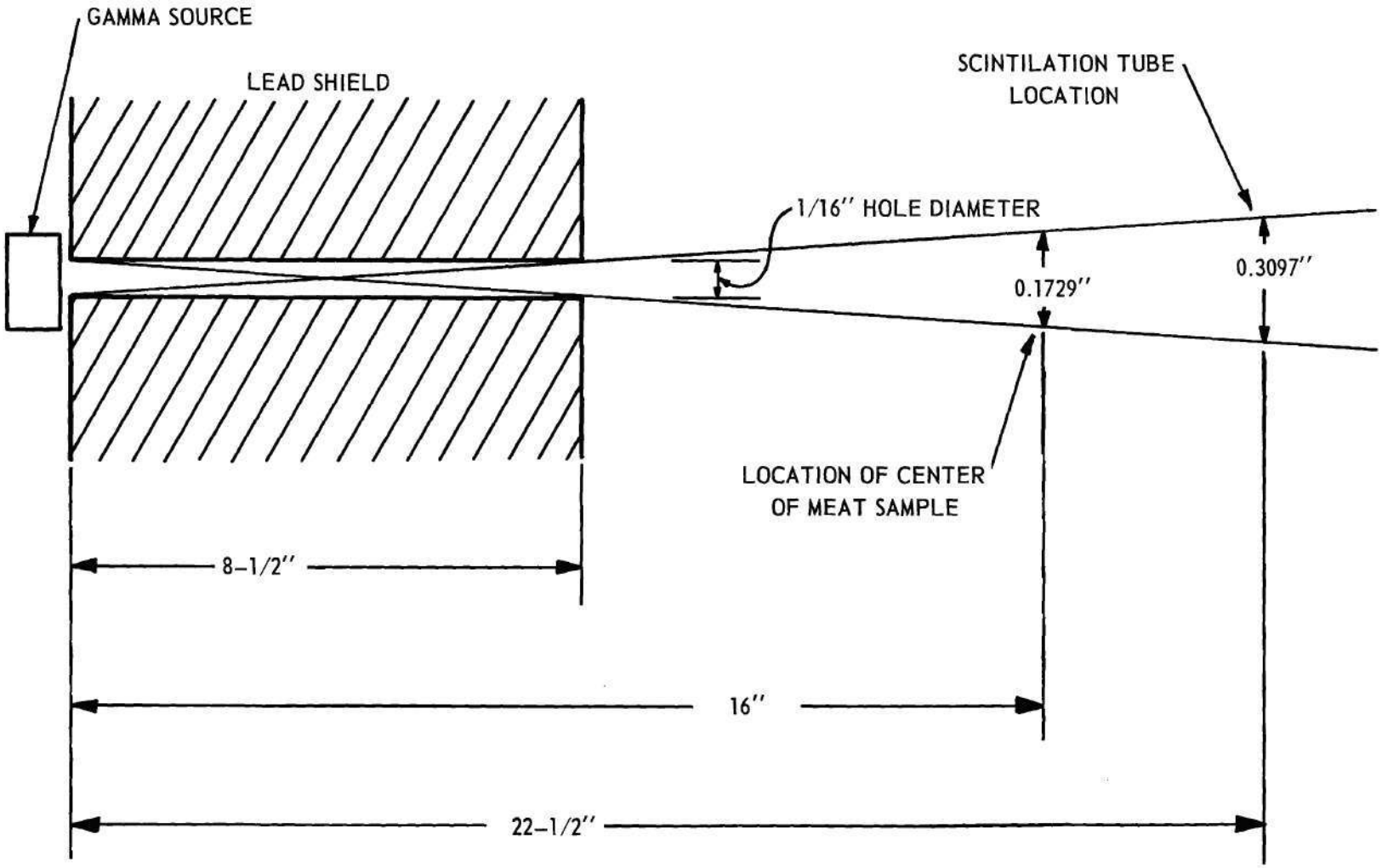

Figure 17. Radiation Beam Geometry.

APPENDIX B

ESTIMATED ERRORS

The estimation of the error for various instruments used in this work is listed in Table 1.

Table 1. Instrumentation Error

Type Instrument	Percentage Error
Wallace-Tiernan pressure gage	2.0
Thermocouples (all)	1.0
Toledo scale	2.5
Cathotometer (including positioning of sample)	4.0
Scintillation Counting	3.3

An inherent limitation of the accuracy of the gamma-ray method to determine moisture content lies in the nature of gamma radiation emission. Although the average emission rate of the cobalt source may be predicted by the half life of the material, the rate at any instant is subject to fluctuations about a mean value due to the random nature of gamma-ray emission from the cobalt. As the events of emission become larger, the distribution of the measured emission rate approaches the Poisson distribution (10), in which the standard deviation of the emission rate is equal

to the square root of the emission rate. In this work the three million counts obtained through the sample constitutes one datum point. Thus, one standard deviation of a single counting period is 1700 counts with a confidence level of 66.6 percent. That is, due to the random nature of radioactive disintegration, a count of three million over a set time period will not be in error by more than ± 1700 counts 66.6 percent of the time.

In order to increase the confidence level, the number of standard deviations must also be increased. To have a confidence level of 99.73 percent requires three standard deviations (10). Therefore, in this work the standard deviation is ± 5100 counts with a confidence level of 99.73 percent. This means that any particular count may be in error by a maximum of 10,200 counts. The difference between the number of counts obtained from a wet and dry sample in this work was approximately one million, which leads to an error in the moisture content determination of 1.0 percent. This error is a result of statistical variation alone and is added to more common errors introduced by counting circuit nonlinearities, timers and relays. The common errors are reduced by good design practices but the statistical error is only reduced by increasing the count rate.

By similar reasoning the statistical error obtained, when using the shorter counting time of 100 seconds for the three millimeter test, was 1.5 percent.

It should be pointed out that much work was done to keep the statistical error as low as possible by increasing the source size and using long counting periods. However, it was found that the variation in count

rate was affected more by instabilities in the scintillation equipment. These instabilities were a result of the high flux of radiation in the beam, thus by obtaining scintillation equipment that would handle high count rates, the accuracy of the system could be improved.

A check was made on the loss in count rate resulting from the natural decay of the cobalt source. Due to the long half-life of cobalt, it was found that during the test period the change in count due to cobalt decay was negligible.

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